An Investigation on the Irradiation Behavior of Atomized U-Mo/Al Dispersion Rod Fuels

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Abstract

The second irradiation fuel experiment, KOMO-2, for the qualification test of atomized U-Mo dispersion rod fuels with U-loadings of 4-4.5 gU/cc at KAERI was finished after an irradiation up to 70 at% U^{235} peak burn-up and subjected to the IMEF(Irradiation material Examination Facility) for a post-irradiation analysis in order to understand the fuel irradiation performance of the U-Mo dispersion fuel. Current results for PIE of KOMO-2 revealed that the U-Mo/Al dispersion fuel rods exhibited a sound performance without any break-away swelling, but most of the fuel rods irradiated at a high linear power showed an extensive formation of the interaction phase between the U-Mo particle and the Al matrix. In this paper, the analysis of the PIE results, which focused on the diffusion related microstructures obtained from the optical and EPMA observations, will be presented in detail. And a thermal modeling will be carried out to calculate the temperature of the fuel rod during an irradiation.

1. Introduction

During the in-reactor operation of a dispersion fuel, interdiffusion or chemical reactions between the fuel particles and matrix occur[1]. Intermetallic compounds in the form of UAl_x are formed as a result of the diffusion reaction. Because the uranium aluminides are less dense than the combined reactants, the volume of the fuel meat increases after the reaction. In addition to the effect on the swelling performance, the reaction layers between the U-Mo and Al matrix induces a degradation of the thermal properties of the U-Mo/Al dispersion fuels[2]. It is important to investigate the thermal behavior of U-Mo/Al dispersion fuel according to reaction between the fuel particles and the matrix with the burnup and linear power.

The first irradiation test(KOMO-1) in HANARO by KAERI for atomized U-Mo dispersion rod fuels revealed the fuels with U-loading of 6.0 g-U/cc to be not acceptable due to the complete interaction between U-Mo particles and Al matrix[3]. In the 2nd irradiation test(KOMO-2) lower U-density fuels with 4.0 and 4.5 g-U/cc were prepared in order to reduce the fuel temperature.

The second irradiation fuel experiment, KOMO-2, for the qualification test of the atomized U-Mo dispersion rod fuels in KAERI was finished after irradiation up to 70 at% U^{235} peak burn-up and subjected to the IMEF(Irradiation material Examination Facility) for a post-irradiation analysis in order to understand the fuel irradiation performance of the U-Mo dispersion fuel[4].

Metallographic examination of the KOMO-2 test fuel rods by the optical observation has been completed recently [4]. Because of the fuels irradiated up to a high burn-up became very brittle in nature, it was difficult to prepare the irradiated sample for the EPMA. Therefore, an examination of the EPMA(Electron Prove Micro Analysis) of several samples with different local burn-ups and sample locations for the 4 g-U/cc U-7Mo/Al fuel(494-L2) has been conducted recently.

In this paper, the analysis of the PIE results, which focused on the diffusion related microstructures, recently obtained from the optical and EPMA observations, will be presented in detail. And a thermal modeling will be carried out to calculate the temperature of the fuel rod during an irradiation.

2. Results and Discussion

Fig. 1 shows the metallographic cross sections, from the fuel meat center to the periphery, of the 4.0 g-U/cc U-7Mo /Al dispersion fuel meat (494-L2) with the burn-ups of 50 % BU, 62% BU, and 68 % BU, respectively. These three fuels were irradiated at the top, middle, and bottom sections of the fuel rod with different BOL(beginning-of-life) temperatures. The same optical presentation for the 4.5 g-U/cc U-7Mo/Al dispersion fuel meat (494-H2) is also shown in Fig. 2. At the 50% BU, the local peak BOL temperatures were 145.5^{o}C for 494-L2 and 134.2^{o}C for 494-H2, respectively. Fuels at 50% BU appeared to exhibit sound irradiation performance due to lower BOL temperature, as expected, although interaction layers seem to develop at the fuel meat center by an interdiffusion of the U-Mo and Al. As the irradiation burn-up increases up to 62% BU and 68% BU, with higher BOL temperature, however, almost all of the U-Mo particles and Al matrix react into the uranium aluminide phase with the remaining very small fraction of pore-like defect, the volume fraction was measured to be about 1.9 %, in which the microstructure at the fuel center(494-H2) seems to be similar to the UO_2, also the dispersion fuel having interaction layers can only be seen at the periphery. In the case of the 4 gU/cc fuel(494-L2) at a high BU, a considerable amount of pore-like defects was observed mostly at the full interaction region in the fuel meat. The volume fraction of the defect was measured to be 9.5%. But in the periphery area of both fuels, no pore-like defect was observed even after 68% BU. However, it is of importance to note that there is no break-away swelling in the rod type U-Mo/Al dispersion fuel although there is a considerable amount of pore-like defects and/or an extensive interaction between the fuel particle and Al.

The interaction layer thickness of the fuel particles was measured along the radial direction and is represented in Fig. 3. The 50% BU samples appeared to have a layer thickness of about 25~35 μm at the center and 7~12 μm at the periphery due to a temperature gradient along the radial direction.(see Fig. 3) In the case of the fuels with a higher BOL temperature, 62% BU and 68% BU, only a few of the unreacted U-Mo particles at the periphery can be seen due to a full reaction of the U-Mo particle with Al, and the thickness of the interaction layer increased rapidly and was measured to be up to ~40-60 μm even at the periphery region. As previously reported, the fuel meat swelling of the rod type U-Mo/Al fuel appeared to be more proportional to the linear power than the U-235 burnup[4], thus, it is mostly dominated by the formation of the interaction phase.

Figures 4-7 represent the scanning electron microscopy of the 494-L2 meat fracture surface from different temperatures and irradiated burnups. At the center of the fuel meat irradiated to 50% BU(Fig. 4), the microstructure of the fuel particle shows a typical low temperature irradiation behavior, similar to the RERTR experiment[5-6]. Small fission gas bubbles in the unreacted U-Mo have been formed at the recrystallized grain boundaries. However, a trace of fission gas bubbles along the grain boundary is also visible in the interaction phase. This phenomenon is attributed to an enhanced formation of the interaction phase having low thermal conductivity, its formation increases the fuel meat temperature, which allows the interaction phase to continue to grow even after recrystallization take place at about 30~40% BU. The BSE(Back Scattered Electron) observation(Fig. 4(f)) indicates that no compositional difference appeared to exist in the interaction phase. Fig. 5 is the fractured surface SEM images from the middle-to periphery of the fuel meat irradiated to 68% BU, e.g., taken at a location representing an on-going full reaction between the U-Mo fuel and Al matrix, in which sample (b) was at the periphery(lower temperature) side and sample (f) was at the meat center(higher temperature) side, respectively. Some of the unreacted aluminum remains between the interlayer-formed fuel particles without any signal of a pore formation. Fission gas bubbles in the unreacted U-Mo (sample (b)) irradiated under a rather lower temperature, remains uniform implying a stable swelling behavior of the fuel. A trace of fission gas bubbles in the interaction phase with a thickness of about 40 μm is also observed, but its size and population is larger than that in Fig. 4. As the fuel temperature increases, however, it is of significance to note that there seems to exist two different reaction layer layers, the inner and outer layers. The outer layer, formed at the initial stage of an irradiation under a relatively low temperature, exhibits no bubbles inside representing a stable swelling behavior.

In the inner reaction layer, many bubbles were precipitated along the recrystallized grain boundaries with the remaining bubble free interaction islands that are suspected of having different chemical compositions with the outer layer. Moreover, the BSE observations of the two layered morphologies appeared to exhibit the same phase.

Characterization of the reaction layer was also carried out by an EMPA analysis for the 4 g-U/cc fuels (494-L2) with different burnup and location, as shown in Fig.8. Similarly to the previous out-of-pile experiment result[7], it is certain that the reaction layer tends to have a composition of $(U,Mo)Al_4$ when the irradiation temperature is relatively low. $(U,Mo)Al_3$ structured phase was identified, even though at both inner and outer layers, for the fuel irradiated at the higher temperature condition. The formation of $(U,Mo)Al_3$ phase in the fully reacted fuel might be also caused by the change in the boundary condition of diffusion process because of the consumption of both the unreacted U-Mo and Al matrix.

The two layered microstructure at the fuel meat center appeared to become diminish as the irradiation of the fuel was progressed at the higher temperature (see Fig. 6). There are also weak traces of fission gas bubbles in the fuel particle center, implying that an extensive interdiffusion reaction due to very high temperature had already finished at a lower burnup.

According to the mass balance calculation, if interdiffusion reaction product is assumed to be $(U,Mo)Al_3$, all the U-7Mo/Al dispersion fuel with a fuel loading of 4.5 gU/cc should react into the uranium aluminide phase resulting in a very small volume change because the apparent density of 4.5 gU/cc fuel meat(6.79 g/cm^3) is closely similar to that of UAl_3(6.8 g/cm^3). When considering the dispersion fuel of 4 gU/cc, there should be a volume increase of about 5 % due to a full interdiffusion reaction and remain the matrix Al of 18.2 vol%. These calculations are coincident with the measured volume fraction data of aluminum including a porosity at the fully reacted fuel zone irradiated to a high burnup, e.g., the dominant phase in the interdiffusion layer formed in U-Mo/Al dispersion fuel should be $(U,Mo)Al_3$ phase. However, the apparent swelling measured from the meat irradiated to 68% BU was to be 15.6% for 4 gU/cc and 11.2% for 4.5 gU/cc, respectively, implying that there is still a considerable difference in the volume change. This discrepancy, about 10-11% in volume, could account for the swelling by an accumulation of fission products in the interaction phase as well as an accumulation of solid and fission products in the U-Mo alloy particles[5].

Because the interaction phase formation rate is strongly temperature dependent and the main effect of the interaction product is the reduction of the thermal conductivity of the fuel meat, it is of significance to establish the maximum temperature of fuel meat under an irradiation condition. The 2nd irradiation test, KOMO-2, for the rod type U-Mo/Al dispersion fuels with U loadings of 4-4.5 gU/cc showed a sound irradiation behavior without break-away swelling, in general, but exhibits a very extensive formation of the interaction phase at the fuel meats with BOL temperatures of above ~160^{o}C. However, there is no available data to know the relation of the interaction phase formation with burnup, it is very difficult to predict a detailed temperature history of the irradiated fuel, because the fraction of the interaction product as well as the linear power changes as the burnup increases. There is also a temperature gradient along the radial direction of the fuel meat, the extent of the interlayer formation can vary widely, and thus it is rather difficult to predict the progress of a real temperature.

Therefore, in this study, we recalculated the BOL temperatures of the fuel meat with the power history under the assumption of no interlayer formation during irradiation, that means the lower limit of fuel temperature[8]. The maximum temperature profiles of the fuel meat were also calculated by assuming that the fraction of the interaction phase is the same as the fraction at the end of an irradiation. By applying this concept, we modified the thermal conductivity of the fuel meat at the end-of-life by dividing the fuel meat into multiple rings, after measuring the volume fractions of each phase at the end of an irradiation. Thus the peak temperature history with the burnup was calculated. Figure 8 represents the potential temperature changes of the 4.5 gU/cc fuel (494-H2) as functions of the burnup and the radial distance from the meat center. This result shows the initial and final temperatures that the fuel meats have experienced during an irradiation period. The peak temperatures of the fuels at the final burnup was calculated to be 143 ^{o}C (50% BU, 134.2^{o}C BOL), 280 ^{o}C (62% BU, 167.3^{o}C BOL), and 334 ^{o}C (68% BU, 188.9^{o}C BOL), respectively. Therefore, the fuel meat having a peak BOL temperature above ~170^{o}C appeared to exhibit an extensive reaction. Moreover, as shown in Fig.8(d) of the temperature distribution of irradiated fuels along the radial direction, it is also found that the full reaction zone corresponds to the radius of the dispersion fuel meat that has irradiated at the final temperature above ~170^{o}C.

From the above result, it is thought that there should be some critical temperature to accelerate the diffusion reaction to make fuel meat covered by the interaction phase in the U-Mo/Al fuel under an irradiation condition. However, the real peak temperature of the fuel meat as the burnup increases is difficult to predict because the growth rate of the interaction layer is dependant on many variables such as U-loading, Mo content, temperature, fission rate, BU, etc.

Recently, we have launched to develop thermal modeling by using FEM in the rod type U-Mo/Al fuel in order to predict the temperature change of fuel meat during irradiation due to the formation of the interaction phase, in which both the interlayer growth kinetics at different fuel meat locations as well as the reduction of the thermal conductivity with burnup will be also considered.[8]

3. Summary

The 2nd irradiation test, KOMO-2, for rod type U-Mo/Al dispersion fuels with U loadings of 4-4.5 gU/cc showed a sound irradiation behavior without any break-away swelling, in general, but exhibits very extensive formation of interaction phase in the fuel meats with BOL temperatures of above ~170^{o}C. The swelling of the rod type U-Mo/Al fuel appeared to be more proportional to the linear power than the U-235 burnup which is mostly dominated by the formation of the interaction phase. (U,Mo)Al_4 phase appeared to form dominantly at the lower temperature region. But (U,Mo)Al_3 structured phase was identified at the interlayers, both inner and outer layers, formed at the higher temperature. The fuel meat having a peak BOL temperature above ~170^{o}C appeared to exhibit an extensive reaction. From the above result, it is thought that there should be some critical temperature to accelerate diffusion reaction to make fuel meat to be covered by interaction phase in the U-Mo/Al fuel under irradiation condition.

4. Acknowledgements

This work has been performed under the nuclear R&D support by the Ministry of Science and Technology, Korea.

5. References

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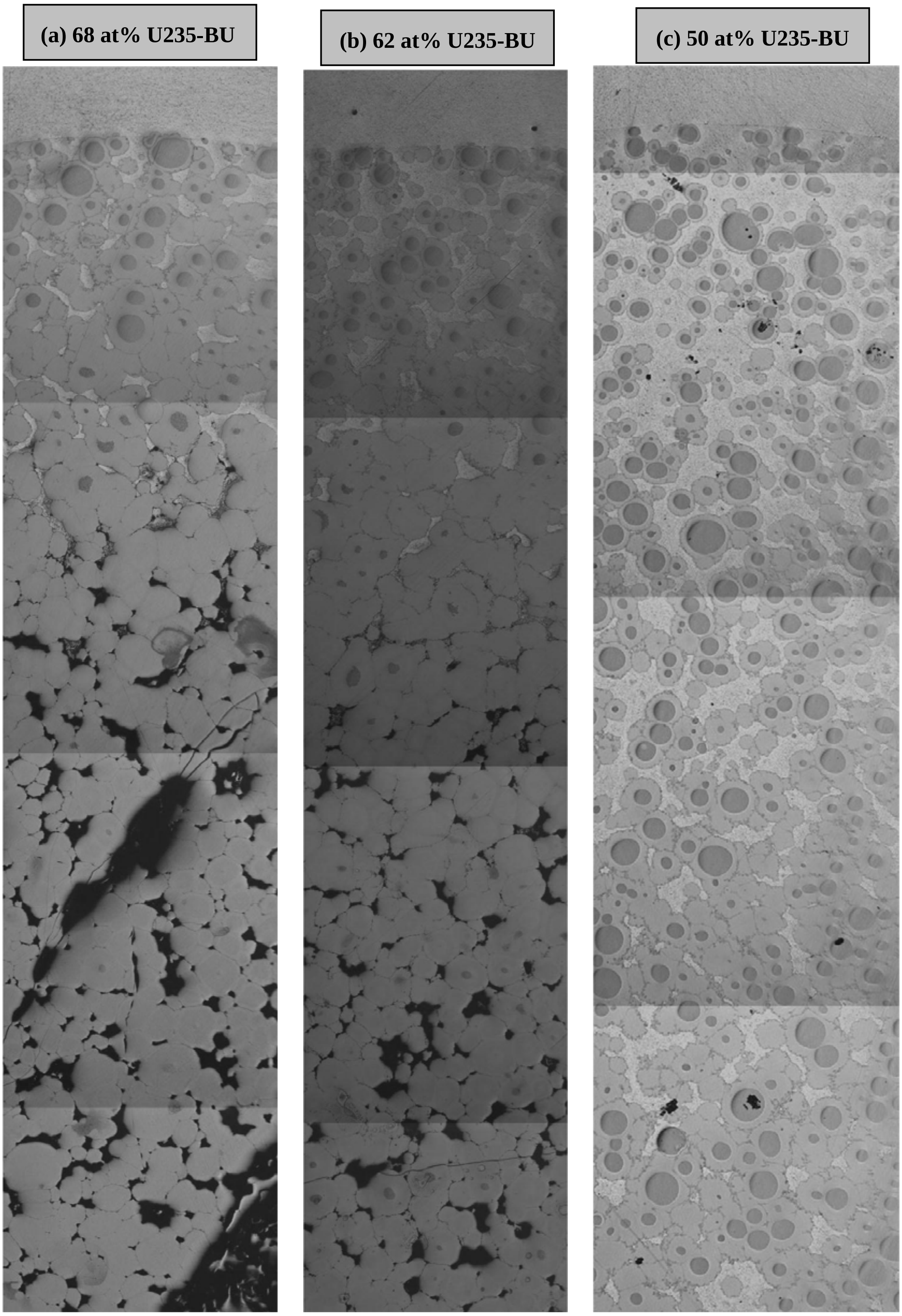


Fig. 1. The metallographic cross sections, from the fuel meat center to the periphery, of 4.0 g-U/cc U-7Mo /Al dispersion fuel meat (494-L2)

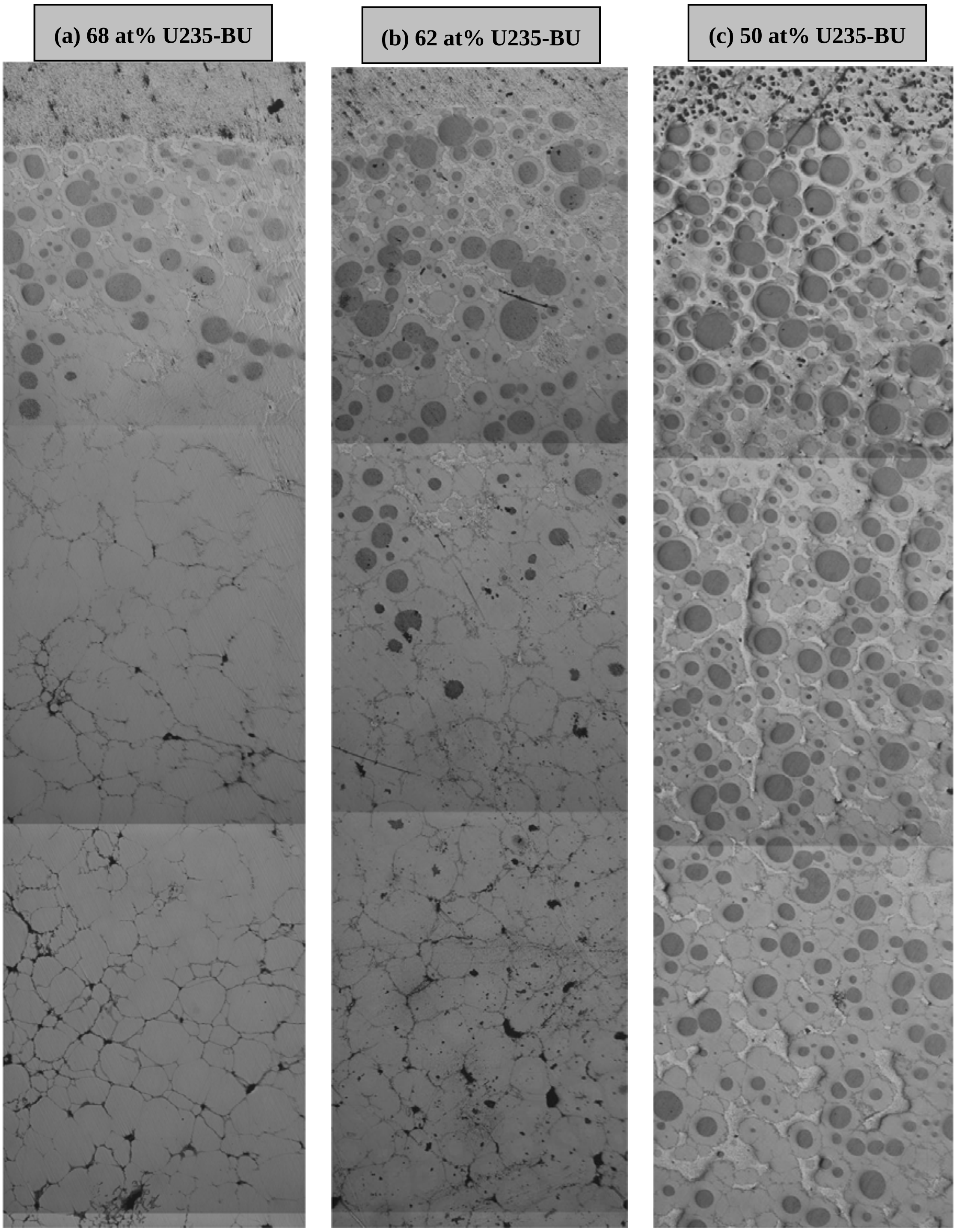
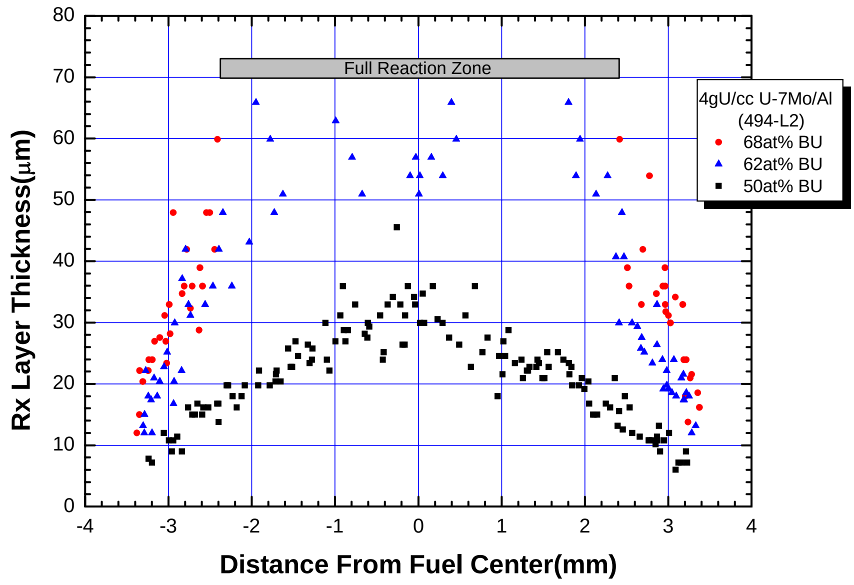
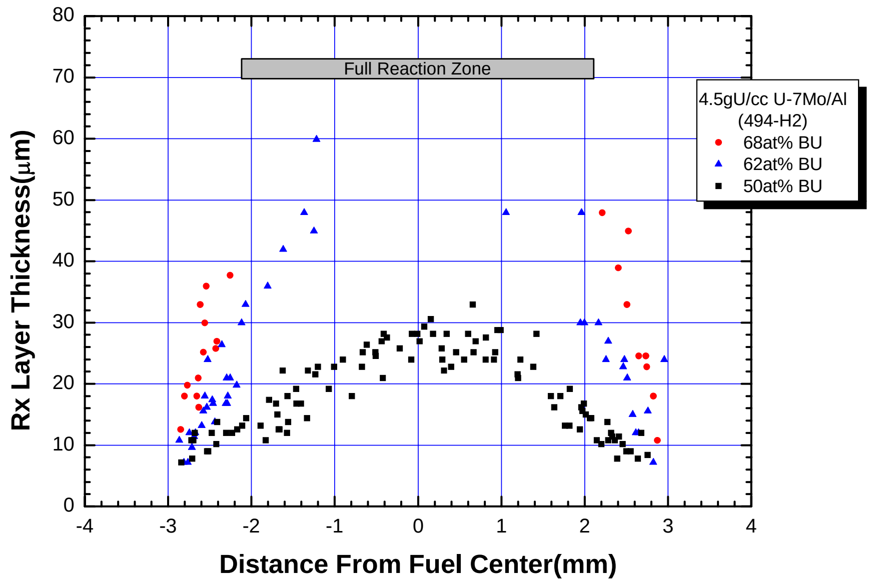


Fig. 2. The metallographic cross sections, from the fuel meat center to the periphery, of 4.5 g-U/cc U-7Mo /Al dispersion fuel meat (494-H2)



(a) 4 gU/cc U-7Mo/Al(494-L2)



(b) 4.5 gU/cc U-7Mo/Al(494-H2)

Fig. 3. Variation of measured interaction layer thickness depending on the radial distance in the fuel meat.

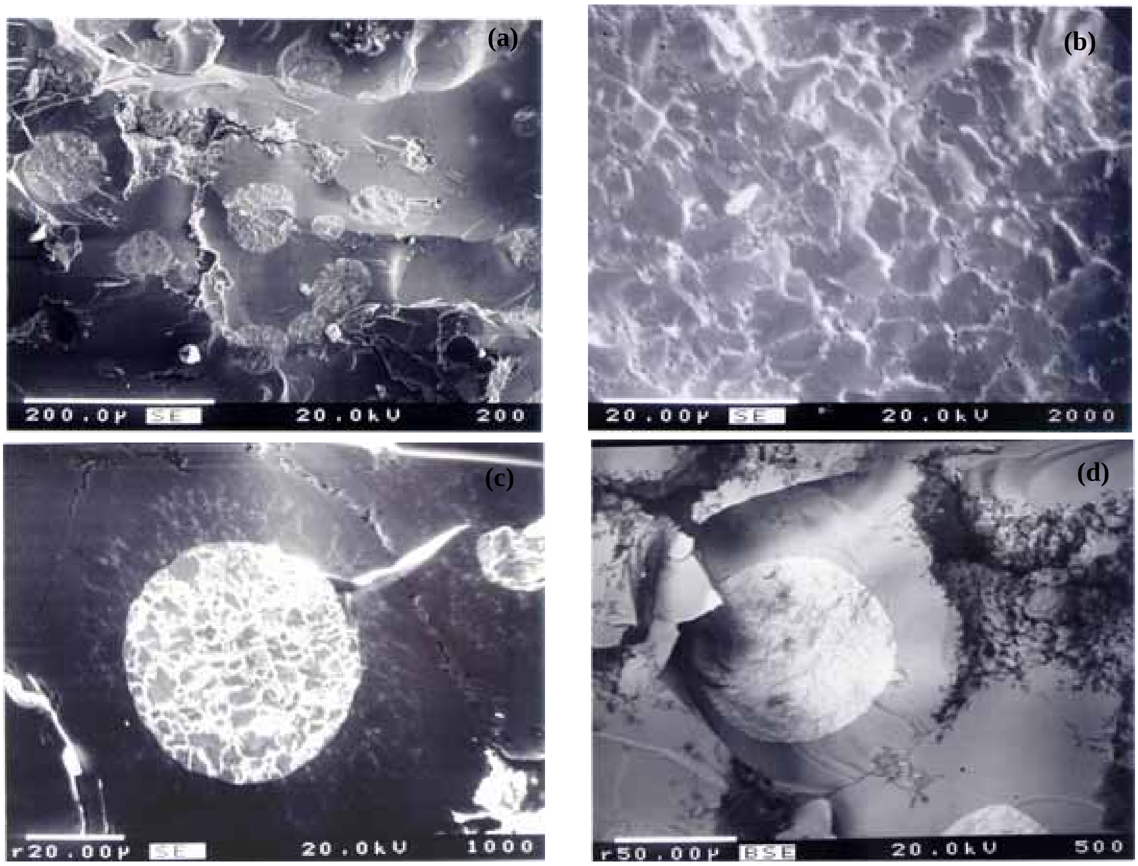


Fig. 4. SEM fractographs of U-7Mo/Al (494-L2) irradiated to 50% BU taken at the fuel meat center.

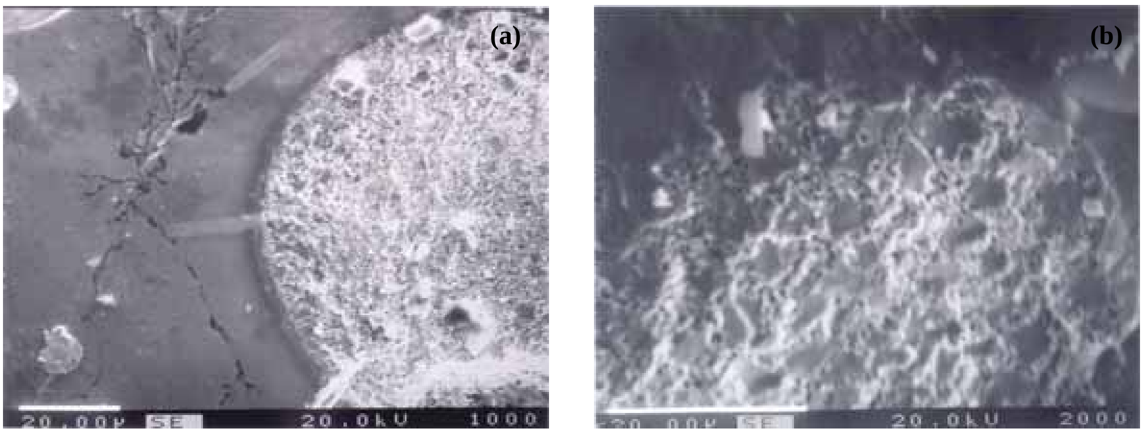


Fig. 5. SEM fractographs of U-7Mo/Al (494-L2) irradiated to 62% BU taken at the fuel meat middle-to-periphery.

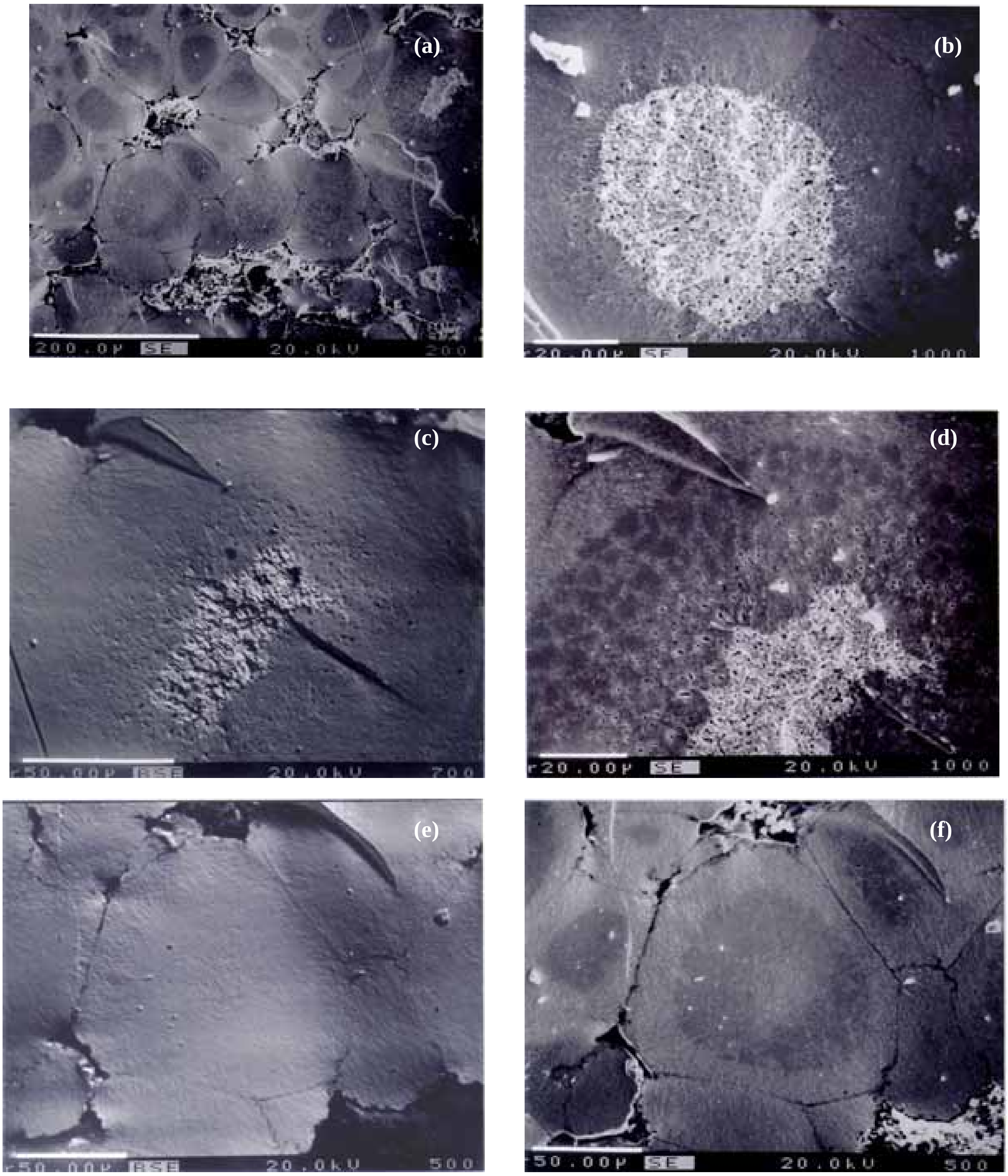


Fig. 6. SEM fractographs of U-7Mo/Al (494-L2) irradiated to 68% BU taken at the fuel meat middle-to-periphery.

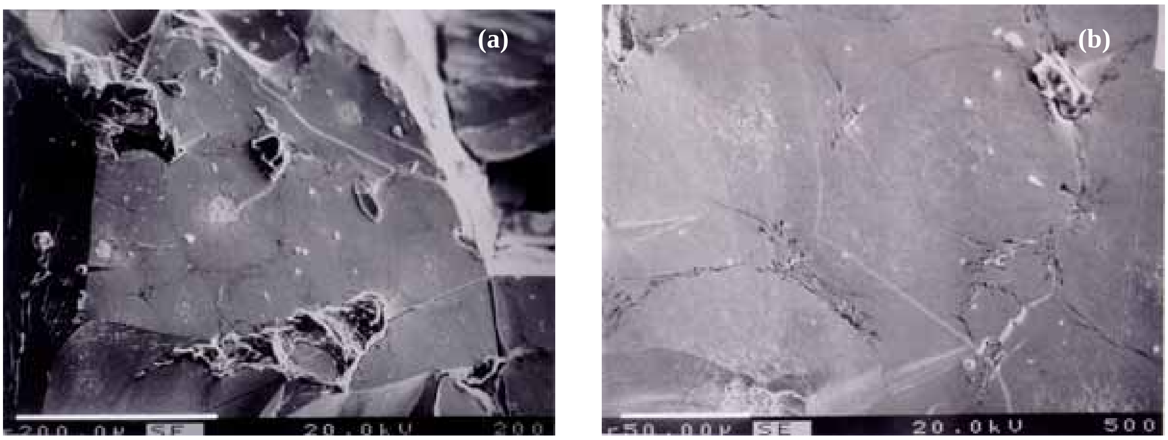
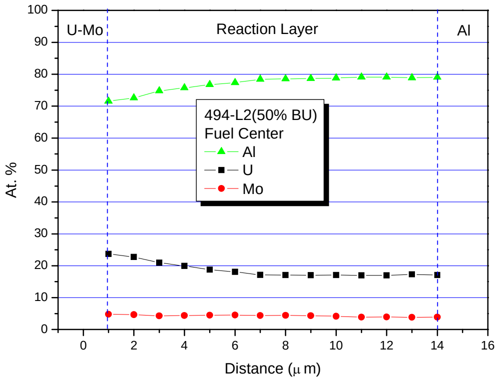
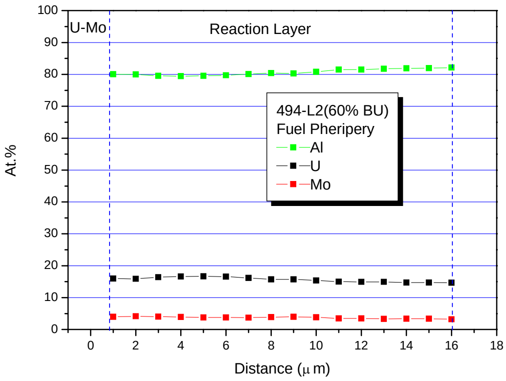
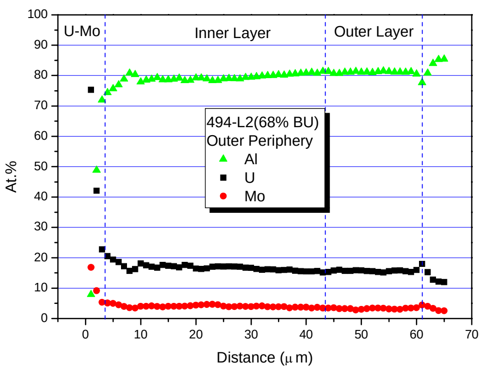


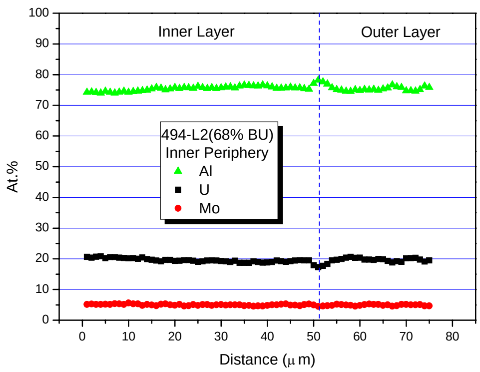
Fig. 7. SEM fractographs of U-7Mo/Al (494-L2) irradiated to 68% BU taken at the fuel meat center.





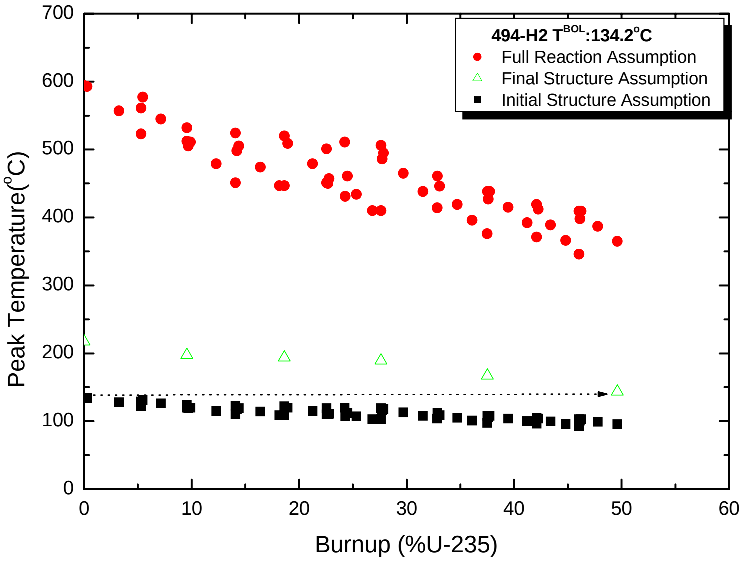
(a) 50% BU, Fuel Center **(b) 62% BU, Fuel Periphery**



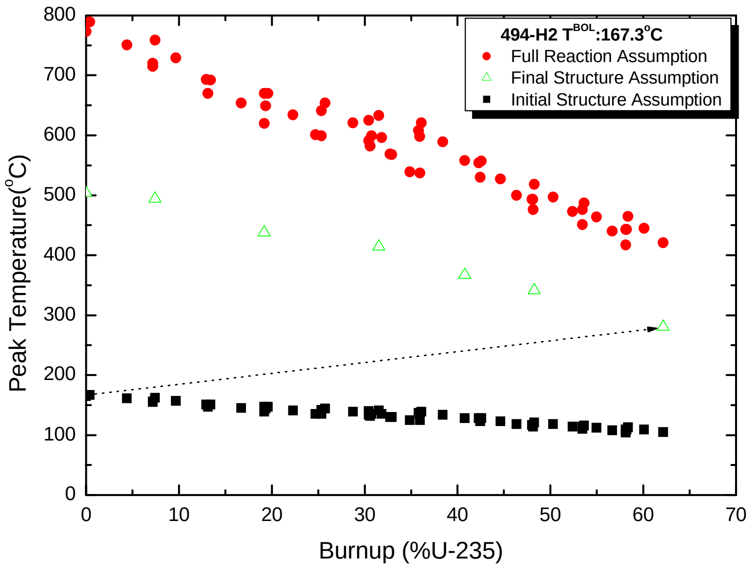


(c) 68% BU, Fuel Inner Periphery **(d) 68% BU, Fuel Outer Periphery**

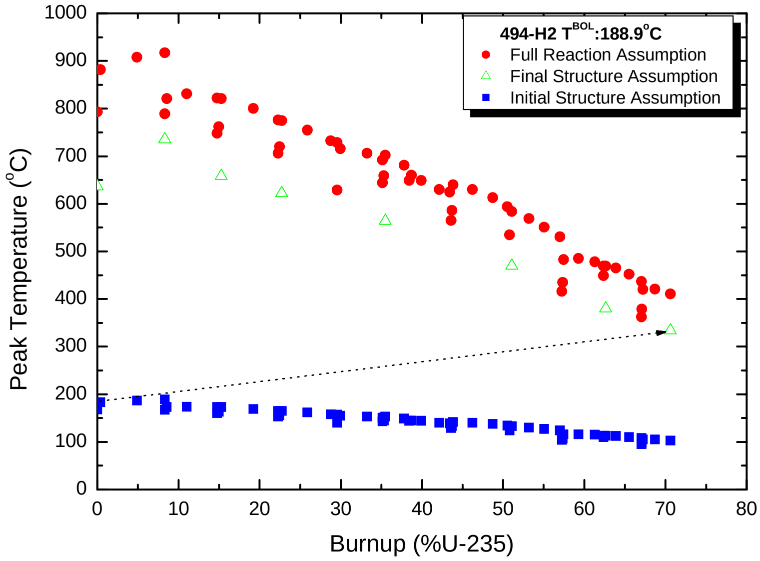
Fig. 8. EPMA scanning results of the interaction layers formed from the fuel particles of (a) Fig.4(c), (b) Fig.5(a), (c) Fig.6(d), and (d) Fig.6(f).



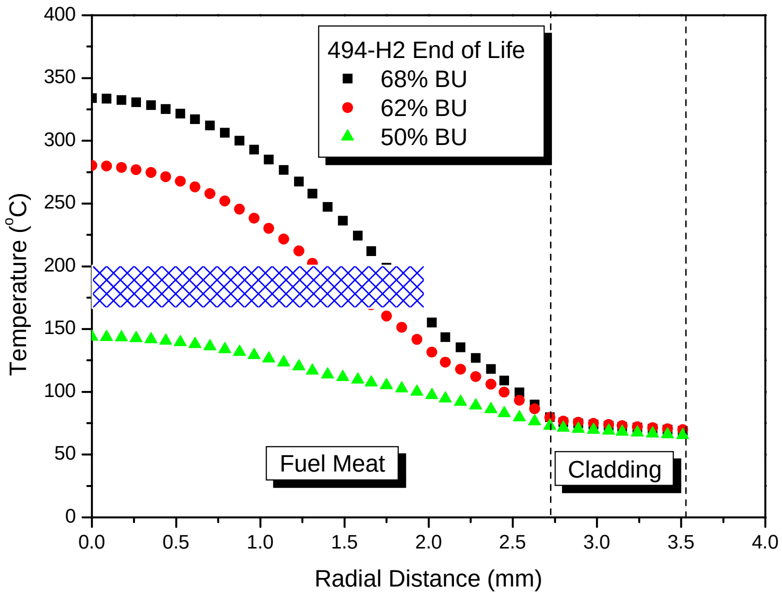
(a) 50% BU (T^{BOL}:134.2^{o}C)



(b) 62% BU (T^{BOL}:167.3^{o}C)



(c) 68% BU(T^{BOL}:188.9°C)



(d) Radial Distribution

Fig. 8. Calculated temperature history of the 4.5 gU/cc U-Mo/Al fuel meat(494-H2).